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Decay Rate and Resonance Fluorescence Spectrum of a Molecule
Near a Composite Material Surface



by

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DECAY RATE AND RESONANCE FLUORESCENCE SPECTRUM OF A MOLECULE NEAR
A COMPOSITE MATERIAL SURFACE

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Abstract

The spontaneous decay rate and resonance fluorescence spectrum of a molecule adapted near the surface of composite materials are calculated in the effective medium approximation. Two substrates are considered: One is a semiconductor containing randomly distributed small dielectric particles and the other a dielectric with metallic particles. Results are analyzed in terms of energy transfer from the molecule to the substrate as functions of the volume fraction of impurity particles.

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I. Introduction

Optical properties, both linear and nonlinear, of composite materials with small solid particles distributed randomly in an otherwise homogeneous medium have been a subject of extensive research in recent years.¹⁻⁵ We consider in this Letter the situation in which a molecule is located near the surface of a composite material. The material can be a semiconductor with small dielectric particles randomly distributed throughout its volume, or a dielectric containing small metallic particles. Our primary aim is to study how the volume fraction of the small particles affect the frequency dependence of optical properties of the adsorbed molecule, in particular, its spontaneous decay rate and light scattering spectrum. As a matter of fact, problems of this sort of adapted molecule have attracted much attention after the fatty-acid monolayer assembly technique⁶ was developed to measure the luminescent lifetime of excited molecules near bulk solid surfaces. As has been shown in previous works on various materials,⁷⁻¹⁰ optical properties of adapted molecules are greatly affected by the structure of the substrate. Therefore, the adapted molecule serves as a probe to explore the structure of the substrate near its surface.

II. Model

Consider a molecule with two levels $|+\rangle$ and $|-\rangle$ separated by a distance $\hbar\omega$ and adsorbed near the surface of a composite material. The molecule is driven by a monochromatic laser field with

$$E(t) = \frac{1}{2} (E e^{i\omega_L t} + E^* e^{-i\omega_L t}) . \quad (1)$$

We can apply the surface-dressed optical Bloch equations (SBE)^{8,9} to describe the interaction process, which are given by⁹

$$\frac{d}{dt} \begin{bmatrix} \langle S^+ \rangle \\ \langle S^z \rangle \\ \langle S^- \rangle \end{bmatrix} = \begin{bmatrix} i(\Delta + \Omega^S) - \gamma & i\Omega & 0 \\ i\Omega^*/2 & -2\gamma & -i\Omega/2 \\ 0 & -i\Omega^* & -i(\Delta + \Omega^S) - \gamma \end{bmatrix} \begin{bmatrix} \langle S^+ \rangle \\ \langle S^z \rangle \\ \langle S^- \rangle \end{bmatrix} - \begin{bmatrix} 0 \\ \gamma \\ 0 \end{bmatrix} . \quad (2)$$

The notation is as follows. The admolecule with a transition frequency ω is located at a distance d away from the surface of a composite solid. The matrix element of the electric dipole moment operator is denoted by $|p|$, and E and ω_L are the amplitude and frequency of the external laser field, respectively. The detuning is $\Delta = \omega - \omega_L$, and the Rabi frequency is $\Omega = |p|E$. The transition probability is proportional to the projection operators defined by

$$S^+ = |+\rangle\langle -| .$$

$$S^z = \frac{1}{2} (|+\rangle\langle +| - |-\rangle\langle -|) \quad (3)$$

$$S^- = |-\rangle\langle +| .$$

The total decay rate of the admolecule can be written as

$$\gamma = \gamma^0 + \gamma^S , \quad (4)$$

where γ^0 is the decay rate in the absence of the substrate.

$$\gamma^0 = \frac{2}{3} \sqrt{\epsilon_1} |p|^2 \omega^3 / c^3 . \quad (4a)$$

with ϵ_1 denoting the dielectric constant of the medium containing the molecule, and

$$\gamma^s = |p|^2 \operatorname{Im} f(d) \quad (4b)$$

is the decay rate induced by the surface. The frequency shift of the spontaneous radiation due to the surface is

$$\Omega^s = |p|^2 \operatorname{Re} f(d) , \quad (5)$$

and the function $f(d)$ is determined by⁹

$$E_R = |p|f(d)S^- = pf(d) , \quad (6)$$

where E_R is the component of the reflected field \vec{E}_R in the direction of \vec{p} .

It is evident that the reflected electric field E_R depends on the structure and other information of the substrate. However complicated the dependence may be, it is contained in the function $f(d)$ which can be determined by usual procedures as described in Refs. 7 and 9. For definiteness, we limit ourselves to the case in which the dipole moment of the admolecule is perpendicular to the substrate surface. The decay rate and frequency shift of the molecule at a distance d from the surface expressed in the unit γ^0 are given by⁷

$$\gamma^s = -\frac{3}{2} \operatorname{Re} \int_0^\infty \frac{\kappa^3 d\kappa}{\mu_1} \frac{\epsilon_1 \mu_2 - \epsilon_2 \mu_1}{\epsilon_1 \mu_2 + \epsilon_2 \mu_1} e^{2i\mu_1 d} \quad (7)$$

$$\Omega^s = \frac{3}{2} \operatorname{Im} \int_0^\infty \frac{\kappa^3 d\kappa}{\mu_1} \frac{\epsilon_1 \mu_2 - \epsilon_2 \mu_1}{\epsilon_1 \mu_2 + \epsilon_2 \mu_1} e^{2i\mu_1 d} \quad , \quad (8)$$

where ϵ_2 is the effective dielectric constant of the composite substrate, $\mu_1 = \sqrt{1 - \kappa^2}$, $\mu_2 = \sqrt{\epsilon_2/\epsilon_1 - \kappa^2}$ and $d = \sqrt{\epsilon_1} wd/c$.

In the composite substrate, we assume that small particles of spherical shape are randomly distributed throughout its volume. Thus, we can employ the effective-medium approximation^{3,5,11} as used in the study of linear dielectric properties of composites. In this approximation the effective dielectric constant ϵ_2 is determined by the equation

$$(1-P) \frac{\epsilon_a - \epsilon_2}{\epsilon_a + 2\epsilon_2} + P \frac{\epsilon_b - \epsilon_2}{\epsilon_b + 2\epsilon_2} = 0 \quad , \quad (9)$$

where P stands for the volume fraction of the particles with dielectric constant ϵ_b embedded in the substrate characterized by the dielectric constant ϵ_a . In general, the solution to (9) is complex and we are only looking for continuous ϵ_2 with positive imaginary part.

III. Numerical results

Two types of composite substrates in a medium of dielectric constant $\epsilon_1 = 1.5$ are considered below:

A. Insulator/semiconductor substrate

The composite in this case is a semiconductor of dielectric function

$$\epsilon_a(\omega) = \epsilon_\infty + \frac{\omega_0^2}{\omega_1^2 - \omega^2 - i\Gamma\omega} \quad (10)$$

containing small insulator particles with frequency independent dielectric constant³ $\omega_b = 25$. In our numerical calculation, we have adopted the unit $\omega_0 = 1$. Other parameters are $\omega_1 = 0.5$, $\Gamma = 0.08$, $\epsilon_\infty = 9$ and $d = 0.5 \omega$. The dependence of the rate γ of spontaneous decay on the frequency can be calculated from Eqs. (4), (7), (9) and (10). The results for various concentrations of small particles are plotted in Fig. 1.

It has been found that the curve peaks at a frequency corresponding to the minimum of the denominator of the integrand in Eq. (7). It may be of some interest to note from our numerical search that the denominator has only one minimum for each P value. It is known that such a resonant condition of the denominator determines the dispersion relation of the surface polariton.¹² If the polaritons do not decay, then the denominator vanishes, namely $\epsilon_1\mu_2 + \epsilon_2\mu_1 = 0$ for polaritons with infinite lifetime. That the minimum denominator remains finite in the present case is because the surface polariton decays. The origin of this damping is the absorption of the composite medium reflected in the imaginary part of the dielectric constant. Therefore the peak of the spontaneous decay rate is a consequence of the energy transfer from the excited admolecule to surface polaritons of the substrate. The curves in Fig. 1 clearly show that a larger concentration of insulator particles in the medium leads to less energy transfer or stronger damping of the surface polariton.

B. Metal/insulator substrate

We now consider an insulator with dielectric constant $\epsilon_a = 25$ and metallic particles distributed randomly throughout its volume. The dielectric function of the metal is

$$\epsilon_b(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\Gamma_p)} , \quad (11)$$

where ω_p is the plasma frequency and Γ_p the damping constant. We employ units such that $\omega_p = 1$ and take $\Gamma_p = 0.01$ and $d = 0.5\omega$ in the following calculation. Numerical computations similar to the above case have been performed, with the results plotted in Fig. 2. It is observed here that the surface plasmon does not appear until the metallic concentration reaches a certain size. Thus there is no evident energy transfer for small P . According to our calculation, the surface plasmon starts to cause energy transfer when $P \geq 0.6$, as can be seen from the peaks in Fig. 2.

We now turn our attention to the resonance fluorescence spectrum in this case. By making use of the regression theorem for correlation functions,¹³ we find from the SBE the well-known results of the incoherent resonance fluorescence spectrum.^{9,14}

$$\tilde{g}(\nu) = \frac{1}{2} |\Omega|^4 \gamma [D^2 + \frac{1}{2}|\Omega|^2 + 4\gamma^2] / [(\frac{1}{2}|\Omega|^2 + |z|^2)(x^2 + y^2)] , \quad (12)$$

where $D = \nu - \omega_L$, $x = 2\gamma(\frac{1}{2}|\Omega|^2 + |z|^2 - 2D^2)$, $y = D(|\Omega|^2 + |z|^2 + 4\gamma^2 - D^2)$ and $z = \gamma + i(\Delta + \Omega^S)$. The effects of the substrate are reflected through γ and Ω^S . We have found from our numerical study that both the height of the peaks and the position of the sidebands are sensitive to any change of the particle

concentration P when the transition frequency of the admolecule is close to the surface plasmon frequency. The peaks in the spectrum diminish as P increases and eventually disappear when P becomes sufficiently large. Part of our numerical results is shown in Fig. 3. It is seen that the presence of the composite substrate does not change the nature of the spectrum. The ac Stark effect still splits each of the two molecular states equally into four levels, such that the resulting three-peak spectrum is symmetric with respect to the main transition frequency. The spacing between lines is affected by Ω^S while the line widths are influenced by γ^S . Of course, both Ω^S and γ^S depend on the nature of the substrate material.

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Figure Captions

1. Decay rate γ vs frequency for dielectric particles embedded in a semiconductor. The volume fraction is $P = 0$ for the solid line, $P = 0.1$ for the dashed line, $P = 0.4$ for the dotted line, and $P = 1.0$ for the dot-dashed line.
2. Decay rate vs frequency for metal particles distributed in a dielectric medium. The volume fraction is $P = 1.0$ for the solid line, $P = 0.8$ for the dotted line, $P = 0.7$ for the dot-dashed line, and $P = 0.4$ for the dashed line.
3. Resonance fluorescence spectrum for a molecule near the surface of a bulk solid which is composed of a dielectric medium in which metal particles are randomly distributed, with $|\Omega| = 45.0$, $\Delta = -5.0$ and $\omega = 0.82$. The solid line is for $P = 0.4$, and the dot-dashed line is for $P = 0.7$.

$$F_{\text{nc}_y} = 1$$

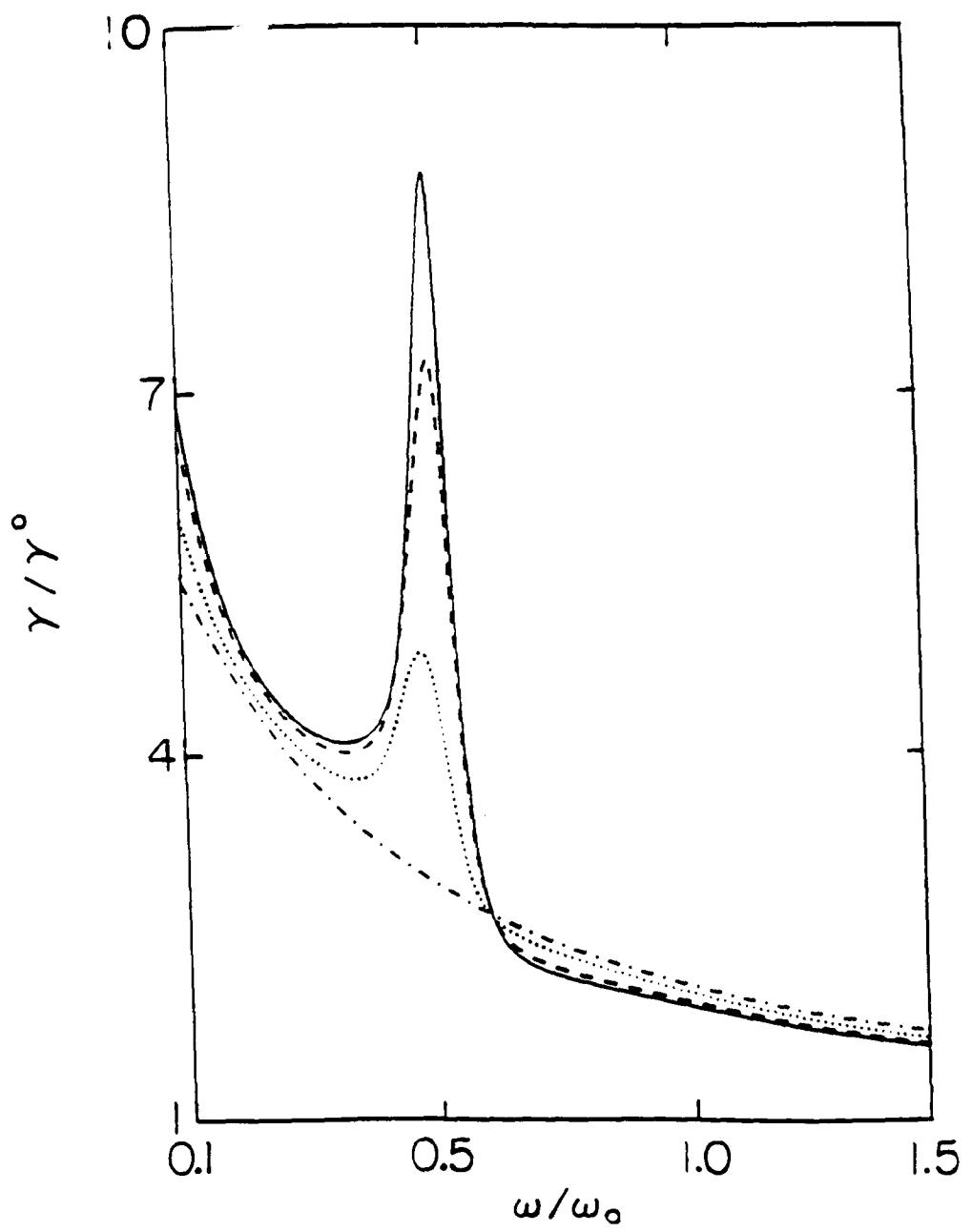


Fig. 2

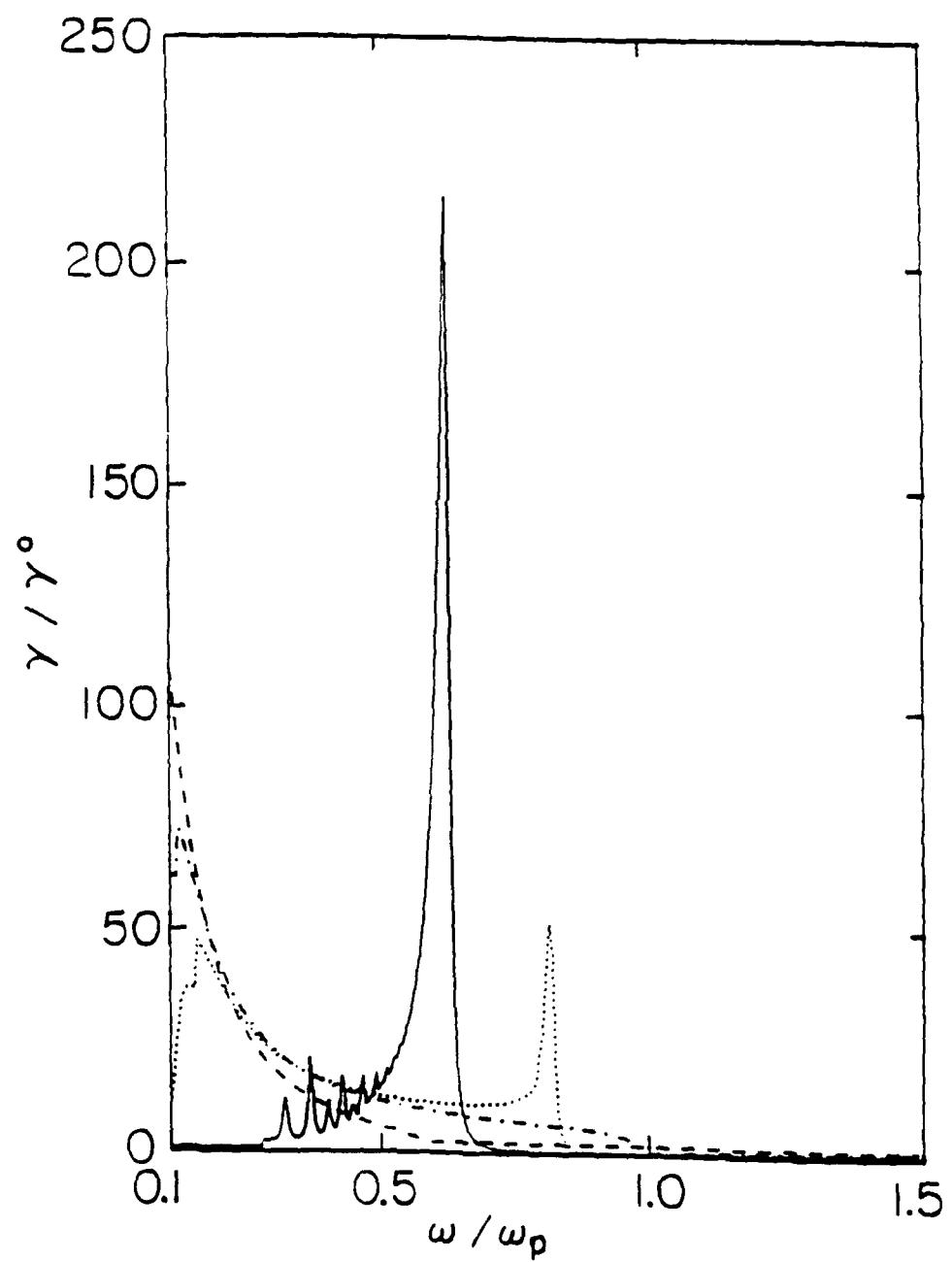
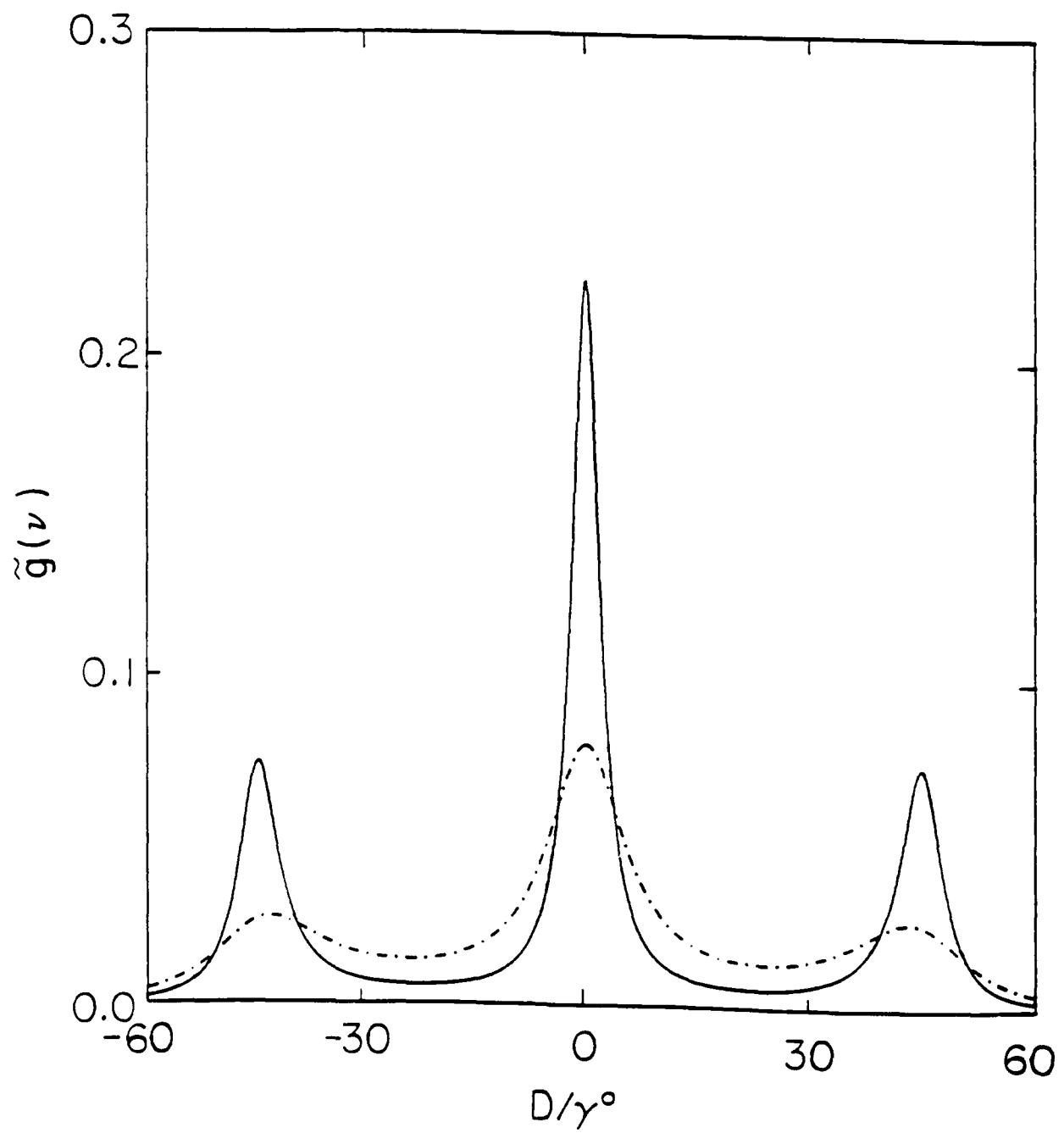


Fig. 3



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